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Exact bound state solutions for sticking of light particles on surfaces

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Abstract : The temperature variation of bound state energies are evaluated for He-LiF system from the pole of exact T-Matrix of the system Green's function. A nice agreement is reported between the calculated and experimental bound state energy for a particular substrate temperature (10^0 K). The temperature variation of the bound state energy is consequently attributed to the inelastic component of the gas–surface scattering.

Keywords : Bound state, Absorption, Inelastic component.

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1. Introduction

Recently there has been renewed experimental and theoretical interest in the low energy sticking probabilities of very light gas particles on physisorbing surfaces[1–3]. The mechanism behind sticking in the quantum regime is understood from the observed sticking coefficients and the elastic scattering probabilities, particularly from a cold surface. However, the attenuation of the elastic intensity of light particles scattered from solid surface seems to be an interesting and important factor in interpreting the sticking mechanism.

In our work, we have theoretically studied the attenuation in the elastic scattering probabilities through the change in BS energy from the change in the pole structure of scattering T- Matrix that corresponds to retaining the real one-phonon process. We have worked out in detail, a quantum statistical theory of a gas particle at the surface of solid system obeying Morse–potential which show localized physisorbtion to a BS, whose energy E_n in magnitude is less than the maximum phonon energy of the solid($\hbar\omega_D$).

2. Theoretical Model

For just one shallow BS the one dimensional static surface potential is

$$V_0(x) = U_0\left(e^{-2\gamma(x-x_0)} - 2e^{-\gamma(x-x_0)}\right) \tag{1}$$

This agrees well with the experimental data though it decreases faster as $x \to \infty$. The model Hamiltonian of the gas solid interaction with the localized and non–localized basis may be written as

$$H = H_{st} + H_s + H_{dyn} \tag{2}$$

where, $H_{st} = \sum_k \epsilon_k n_k + \sum_q \epsilon_q n_q$,
$n_k = c_k^\dagger c_k$, $n_q = \alpha_q^\dagger \alpha_q$ are the occupation numbers for nonlocalized and localized scattering states respectively, and ϵ_k, ϵ_q are the single particle energies in the state k and q. Now using the linear transformation $\alpha_q = \sum_q \phi_q(k) c_q$ for diagonalization we have $H_{st} = \sum_q E_q \alpha_q^\dagger \alpha_q$, where q labels both the BS energy and the continuum energy $E_k = \frac{\hbar^2 k^2}{2m}$ with the gas particle mass m. The $\alpha_q^\dagger$ and α_q are the creation and annihilation operators of a gas particle in the state q. The Hamiltonian of the solid in the harmonic approximation will be

$$H_s = \sum_p \hbar\omega_p b_p^\dagger b_p \tag{3}$$

with $b_p^\dagger$ and b_p are the creation and annihilation operators of longitudinal acoustic phonons of frequency ω_p. The phonon–mediated gas solid interaction is accounted for, by the dynamic part of the Hamiltonian which in lowest order harmonic approximation is given by

$$H_{dyn} = \sum_{q,q-p} \chi(q, q-p)\alpha_{q-p}^\dagger \sum \omega_p^{-\frac{1}{2}}(b_p^\dagger + b_{-p})\alpha_q \ , \tag{4}$$

where for local surface potential we have

$$\chi(q, q') = \left(\frac{\hbar}{2MN}\right)^{-\frac{1}{2}} \int \phi_{q-p}^*(x) \frac{dV_0(x)}{dx} \phi_q(x) dx, \tag{5}$$

where the ϕ_q's are the eigenfunctions of H_{st} *i.e.*,

$$\left(-\frac{\hbar^2}{2m}\frac{d^2}{dx^2} + V_0(x)\right)\phi_q(x) = E_q \phi_q(x) \tag{6}$$

with solid particle mass M and N, the particle number in a normalized box of the gas of length L.

By similarity transformation the Hamiltonian(2) neglecting the phonon absorption is reduced to

$$H = \sum_q E_q \alpha_q^\dagger \alpha_q + \sum_p \hbar\omega_p b_p^\dagger b_p + \frac{1}{2}\sum_{q,q'} \chi(q-p,q)\,\chi(q'+p,q') \times \lambda_p^2 \alpha_{q'+p}^\dagger \alpha_{q'} \alpha_{q-p}^\dagger \alpha_q \left(\frac{1}{E_q - E_{q-p} - \hbar\omega_p}\right) . \quad (7)$$

where $\lambda_p^2 = \frac{1}{\omega_p}$ Now, in order to obtain the Dyson equation for the scattering T–Matrix we write the single particle Greens function as,

$$G_{km}(t) = << \alpha_k(t), \alpha_m^\dagger(0) >> . \quad (8)$$

The Greens function may be written in the form of Dyson equation using the Hamiltonian (7) and taking the Fourier transformation as

$$G_{kk}(E) = G_k(0) + G_k(0) T G_{kh}(E) \quad (9)$$

which on iterative solution leads to

$$G_{kk}(E) = G_0(E) + G_0(E) T G_0(E) \quad (10)$$

where,

$$G_0(E) = \frac{1}{2\pi(E-E_k)},\ T = \left(\frac{2\pi\Delta_k}{1-\sum_q \frac{\Delta_q}{E-E_q}}\right)$$

where,

$$\Delta_q = \sum_p \frac{|\chi(q,q-p)|\lambda_p n_{q-p}}{E_q - E_{q-p} - \hbar\omega_p} .$$

Here, q and (q-p) are the localized state and BS momenta respectively and E is the effective final energy with transformed BS energy E_n due to gas–solid interaction plus a phonon energy.

The relative gas atom occupation number in the substrate maintained at temperature T_s and pressure P is
$n_{q-p} = \exp[\beta(E_n' - \mu)] \quad << 1$
with $\beta = \frac{1}{K_\beta T_s}$ and μ is the chemical potential of gas. Now while evaluating $|\chi(q,n)|^2$ we consider for convenience the dimensionless parameters
$\sigma_0^2 = \frac{2mU_0}{\hbar^2\gamma^2}$, $\xi = \gamma x$, $\xi_0 = \gamma x_0$ and get the normalized BS wave function as

$\phi_n(x) = \sqrt{\gamma} f_n(\xi)$, with

$$f_n(\xi) = (2\sigma_0)^{S_n}\Gamma^{-\frac{1}{2}}(2S_n)\begin{bmatrix} 2S_n + n \\ n \end{bmatrix}^{-\frac{1}{2}} \times \exp\left(-\sigma_0 e^{-(\xi-\xi_0)}\right) e^{-S_n(\xi-\xi_0)} L_n^{2S_n}\left(2\sigma_0 e^{-(\xi-\xi_0)}\right), \quad (11)$$

where $S_n = \sigma_0 - n - \frac{1}{2}$; with n = 0, 1, 2,......... and $L_n^{2S_n}(u)$ is a Laguerre Polynomial.

The continuum state wave function of momentum q normalized in a box of length L, $(-L < x < L)$, are given by
$\phi_q(x) = (2L)^{-\frac{1}{2}} f(\eta;\xi)$, $\eta = q/\gamma$ and

$$f(\xi,\eta) = \mid \frac{\Gamma(1/2-\sigma_0-i\eta)}{\Gamma(2i\eta)} \mid exp\left(-\sigma_0 e^{-(\xi-\xi_0)} e^{-i\eta(\xi-\xi_0)}\right) \psi\left(\tfrac{1}{2} - \sigma_0 + i\eta, 1 + 2i\eta, 2\sigma_0 e^{-(\xi-\xi_0)}\right), \quad (12)$$

where $\psi(a,b,z)$ is a confluent Hpergeometric function that vanishes as $z \to \infty$. Again while evaluating the sums we use the thermodynamic limit *i.e.* $\sum_k \to (\frac{L}{\pi})\int_0^\infty dk$ to perform the sum over the phonon states for a Debye model *i.e.* $\sum_p \to (\frac{3N_s}{\omega_D^3})\int_0^{\omega_D} \omega_p^2 d\omega_p$ This leads to

$$\sum_q \frac{\Delta_q}{(E-E_q)} - \frac{3(2\sigma_0 - 2n - 1)}{8r^3(n!)\Gamma(2\sigma_0 - n)}\frac{m}{M}\exp\left(\frac{E_n-\mu}{K_\beta T_s}\right) \times \int_0^c \frac{\sinh(2\pi\sqrt{x})}{\left(\sin^2(\pi\sqrt{x}) + \cos^2(\pi\sigma_0)\right)} \frac{\left(x + \left(\sigma_0 - n - \frac{1}{2}\right)\right)}{(r + S^2 - S_n^2 + x)} \times \left|\Gamma\left(\frac{1}{2} + \sigma_0 + i\sqrt{x}\right)\right|^2 \left(r + x ln \left|\frac{r-x}{x}\right|\right) dx \;. \quad (13)$$

The equation (13) has been evaluated numerically to obtain the BS solution from the resonance condition *i.e.*

$$1 - \sum \frac{\Delta_q}{E - E_q} = 0 \;. \quad (14)$$

3. Results

Using the temperature independent zeroth BS (E_0) energy from the Morse potential, we have evaluated the temperature dependent part of the BS from the equation (14). For the substrate temperature $T_s = 10^0K$. The Morse

Table 1: Morse potential parameters [4], BS energy and substrate temperature. $\gamma^{-1}(\AA) = 1.09$, $U_0(K) = 89.0$, $r = 145.79$, $\sigma_0 = 4.1979$, $m/M_s = 0.152$, $\hbar\omega_D/K_\beta(K) = 730.0$

BS energy in ^{0} K		Substrate temp in ^{0} K	
Expt. value [5]	Cal. value	Expt. value [6]	Cal. value
68.47	10	68.48	8.2

Table 2: Variation of BS energy with substrate temperature due to phonon mediated inelastic part.

T^0 K	8.25	8.5	8.75	9.0	9.5	10.0	10.25
$E_0'/K_\beta{}^0K$	68.97	71.25	73.53	75.82	80.40	85.0	87.31
$\Delta E = E_0' - E_0$	0.5	2.78	5.06	7.37	11.93	16.53	18.84

potential parameters and the temperature dependent BS energies for different substrate temperatures around 10^0K have been given in Table 1. The substrate temperature evaluated by fitting the experimental BS energies agrees well with the reported value. The component of the BS energy which contribute to the inelastic scattering may be estimated from the calculated results of variation of BS given in Table 2. The nature of variation of BS energy with temperature is shown in Fig. 1.

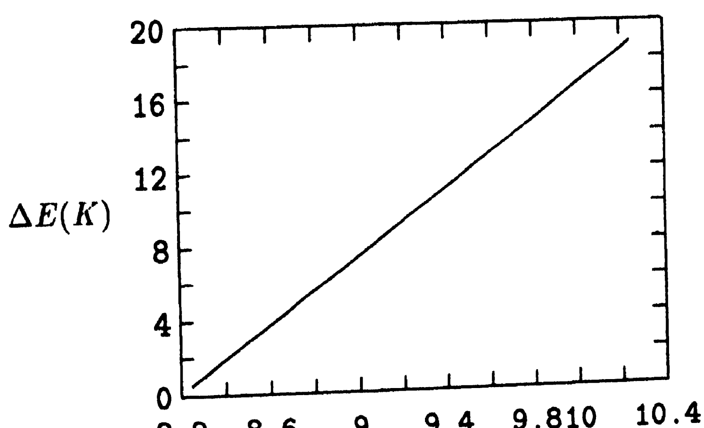

Fig 1. Variation of BS energy($\Delta E(K)$) with substrate temperature(T(K))

3. Conclusion

We have evaluated the change in the BS energy due to phonon mediation for He beam incident on the LiF surface and have shown that the phonon mediated BS energy changes with temperature. This indicates that the probability of capture by a BS increases with slight variation of the substrate temperature T_s. If we increase T_S furthermore the higher order effects become more important and more channels for BS capture become probable. Form the nature of the curve in Fig. 1, it is obvious that the rise in substrate temperature implies that as more and more one phonon energies are emitted the BS energy will go deeper and deeper.

Acknowledgments

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References

[1] S. Anderson et. al. *Phys. Rev.* **40** 8146 (1989).

[2] H. Schlichting et. al. *Phys. Rev. Lett* **60** 2515 (1988).

[3] G. Armand and J. R. Manson *Phys. Rev. B* **43** 14371 (1991).

[4] Z. W. Gortel et. al. *Phys. Rev. B* **22**, 5655 (1980).

[5] G. Derry et. al. *Surface Science***74** 245 (1978)

[6] *Dynamics of Gas Surface Scattering F. O. Goodman and W. H. Y. Watchman* (New York : Academic) (1976)